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THE USE OF MODELS FOR SOURCE APPORTIONMENT AND FOR ASSESSING THE CONTRIBUTION OF NATURAL SOURCES IN RESPONSE TO THE AIR QUALITY DIRECTIVE

Evangelia Fragkou, Ioannis Douros and Nicolas Moussiopoulos

Laboratory of Heat Transfer and Environmental Engineering, Aristotle University of Thessaloniki, Greece

Abstract: The sub-group on the contribution of natural sources and source apportionment has been established within the framework of the Forum for Air Quality Modelling in Europe (FAIRMODE), co-funded by EEA and JRC. This sub-group belongs to the working group on quality assurance of models and focuses on model use for source apportionment and for estimating the contribution of natural sources on pollutant concentrations. The activities within the sub-group, aim to provide useful guidance and suggest best modelling practices and quality assurance procedures for member countries, in order to promote harmonised model use for policy applications. This advice is expected to be of further benefit to member countries, as one of the key elements in the current Air Quality Directive is the possibility to discount natural sources of pollution (especially in the case of PM) when assessing compliance against limit values.

More specifically, a detailed analysis on current modelling practices is undertaken, in order to identify the basic needs for harmonisation. The analysis is primarily based on an extensive review of the ways models are being used for source apportionment in relation to policy purposes by relevant authorities and research groups in member countries, putting special emphasis on methods to quantify uncertainties. An important aspect of this review is presented in this paper and constitutes an analysis of the modelling methods that have been applied by different member countries in preparation of the report for postponement of attainment of PM_{10} limit values. At a later stage, the modelling methods reported in the applications will be compared and evaluated with regard to a number of criteria including accuracy and input data requirements, and a harmonised methodological framework will be suggested for assessing natural contributions. This will provide useful input to the Guidance Document produced by the European Commission Work-Group for Implementation in order to assist member countries on issues related to source apportionment in the framework of the Air Quality Directive.

Key words: *FAIRMODE, source apportionment, PM natural contribution, use of modelling in EU Directive***.**

INTRODUCTION

Pollutant levels exceeding EU limit values for the protection of health and the environment constitute a major concern of most European cities. This is also reflected in the significant number of EU countries that have applied for a postponement of attainment of PM_{10} limits until 2011, according to Article 22 of the Air Quality Directive (AQD) 2008/50/EU. Efficient air quality management is required in order to ensure that the legal limits are not exceeded and that the economical and social costs of poor air quality are controlled and minimised. The first step to ensure the application of successful mitigation measures and the development of appropriate pollution abatement national strategies is the accurate identification of pollution sources and of their individual contributions to the ambient pollutant concentrations. Therefore, the development of a harmonised approach for this process of source apportionment, which could be applied for a range of pollutants, source types and horizontal scales and documented as an easy-to-use guidance, would be of great benefit to the relevant authorities and regulatory bodies in all EU countries.

In response to this need, a working sub-group (SG) on the “Contribution of natural sources and source apportionment” has been formed within the frame of the Forum for Air Quality Modelling in Europe (FAIRMODE). Although not explicitly mentioned in the AQD, the use of modelling tools in combination with measurements is recommended for the purpose of source apportionment, as monitoring of the contributions from all emitting sources in an air quality management zone or agglomeration would be unrealistic and economically unfeasible. Therefore, the main scope of the SG is to provide useful guidance and suggest best modelling practices and quality assurance procedures for member countries, in order to promote harmonised model use for source apportionment in the EU. At a first stage, this will be achieved through an extensive review of the current status of modelling practices used for source attribution and quantification of contributions by member states, in order to identify gaps, limitations and needs for harmonisation. The material used for this review includes information from a relevant database compiled within the frame of the COST Action 633, from a workshop that took place at the JRC premises in Ispra in October 2006 on the “Quantification of the contribution of natural sources to the ambient PM concentrations” and from the analysis of the extension reports submitted by member countries in support of their applications for postponement to comply with PM_{10} limit values. Indicative recent publications from member countries on source apportionment with the use of models are also consulted. This paper focuses on the use of models for source apportionment regarding PM_{10}. Due to the health risks associated with their increased concentrations in a number of epidemiological studies (Pope, C.A. *et al.*, 2002) and the vast variety of their sources, source apportionment of PM_{10} has emerged as an important scientific and management issue.

MODEL USE FOR SOURCE APPORTIONMENT

At a first stage, source apportionment may be based on the observation and analysis of monitoring data, through examining their correlation with relevant meteorological parameters and subtracting of levels at regional background from the urban background and hot-spot concentrations, in order to determine the importance of local sources. A similar methodology can be used to quantify natural contributions, however in this case PM regional background levels are subtracted from those measured at the urban and traffic stations of interest for a specific period of days. The occurrence of concentration peaks of measurements simultaneously at different stations which are representative for a variety of horizontal scales can indicate an episode due to transboundary pollutant transport or due to an accidental release. Although the importance of monitoring data for screening purposes and for an initial air quality assessment cannot be argued, their use is subject to limitations, mainly due to issues of spatial and temporal representativity compromised by the increased costs associated with adequate coverage and reliability. Therefore, source apportionment based solely on the use of monitoring data is usually applied for limited time periods, for which high resolution measurements can be obtained, for example from dedicated campaigns (Gelencsér, A. *et al.*, 2007).

A detailed and accurate source attribution requires the combined use of models and data from monitoring networks. For this purpose, simple statistical receptor models have been traditionally used, however more complex dispersion models have recently been also considered by national authorities of member countries, according to selected publications. Sector zero-out modelling, where model simulations are performed assuming zero pollutants emissions from sources of interest or actual source apportionment modelling for the estimation of contributions from different sources in a single simulation are common modelling methodologies followed.

Receptor models

During the past three decades, receptor models have been widely used for source apportionment. The fundamental principleuof receptor modelling is that mass conservation is assumed and a mass balance analysis can be used to determine and apportion ambient pollutant concentrations to individual emitting sources. A mass balance equation is written to account for all chemical species identified in the filter samples analysed, as contributions from independent sources. Receptor models represent a statistical evaluation of ambient measurements at different times and locations, thus the selection of the appropriate method depends on prior knowledge on the sources and source profiles. If the sources are known and detailed information on source profiles is available, Chemical Mass Balance (CMB) models can be applied, whereas in case the sources are unknown and there is limited information on source profiles, Principal Component Analysis (PCA) and Positive Matrix Factorization (PMF) methods are preferred. Apart from the different Chemical Mass Balance modelling tools, receptor models can also be categorised as Aerosol Evolution and Equilibrium models, which estimate how reduction in one precursor will affect PM end-products, and as Back Trajectory models, which identify the origin of polluted air masses usually transported to the receptor from long distance sources. A review of European publications, which were reported in the questionnaires submitted by EU countries within the frame of the COST 633 Action, revealed that PCA was the most frequently used modelling method for source apportionment, as it appeared in 30% of the studies, while back-trajectory analysis was represented in 11% of the studies (Viana, M. *et al.*, 2008). Other receptor models were also frequently used, such as PMF (8%), CMB (7%) and mass balance analysis (7%). In a relevant study by Pio, C.A. *et al.* (1996), PCA modelling was used to separate and identify the major sources of atmospheric aerosol in the samples collected from a regional background sampling site on the western Portuguese coast.

Dispersion models

In contrast to the receptor models that use ambient concentrations as inputs to calculate source contributions, dispersion models use source emissions as input data to calculate ambient concentrations. Eulerian and Lagrangian dispersion models describe the chemical and physical atmospheric processes in order to predict pollutant concentrations and can be applied at different spatial scales. Gaussian dispersion models are source-oriented models that are useful for source apportionment purposes as they characterise atmospheric processes by dispersing a pollutant directly emitted from the source of interest at selected downwind receptor locations. Gaussian simulations allow for the assessment of contribution from a new source to the ambient pollution load, however multiple sources can also be considered in a single simulation. Recently, several EU countries have relied on the combined use of available emission data and a dispersion model to estimate source contributions. For example, the hybrid Swedish AIRVIRO dispersion model has been applied in a number of European cities including Prague, Riga, Vilnius and Tallinn. The Gaussian ADMS-urban model has been used for source apportionment and for the evaluation of proposed emission reduction measures in Poland, within the frame of an air quality management project in Cracow between 2005 and 2006 (Adamczyk, L. *et al.*, 2007). Eulerian dispersion modelling systems have been used recently in Mediterranean member states to assess the Saharan dust contribution to ambient particle concentrations. In the studies by Astitha, M. *et al.* (2005) and Kallos, G. *et al.* (2006), the SKIRON/ETA dispersion forecasting system was applied for urban Mediterranean regions, while Rodríguez, S. *et al.* (2001) have combined SKIRON results with back-trajectory analysis to determine the proportion of Sahara-induced exceedances with respect to the total annual exceedances in Southern Spain.

Model validation and estimation of uncertainty

The AQD explicitly designates the use of modelling techniques for air quality assessment purposes. Although not explicitly mentioned, the EU official guidelines suggest that models are the scientifically relevant tools to be used also for source apportionment. Thus, the models applied for source apportionment have to be tested and assessed in order to ensure that they meet certain quality objectives before considered to be suitable for regulatory use. In general, model validation is performed by the means of comparing model results to measurements. Monitoring data accuracy and coverage has to be ensured in this case, thus it is recommended that data from dedicated monitoring campaigns are used for this purpose, such as in the study by Simpson, D and K.E. Yttri (2009) for Switzerland, Sweden and Norway. A number of recent European publications focus on the intercomparison of different receptor models applied to a single data set. In most cases there is considerable disagreement between results from different modelling tools, due to the different underlying theoretical assumptions. The recent study by Favez, O. *et al.* (2010) compares the performance of CMB and PMF receptor models in determining the contribution of wood burning organic aerosols to the total organic fraction of the field samples obtained from Grenoble, France. It was found that the CMB model overestimated the wood burning contribution, probably due to the loss of semi-volatile compounds from sources to the receptor site. In this way, model intercomparison can provide useful information on model accuracy and reliability, particularly if model results are also evaluated against measurement data. Thus, the use of more than one receptor modelling approach is a useful validation tool when apportioning sources of the ambient aerosol, revealing model limitations for specific pollutants, spatial scales and applications. Through similar exercises, hybrid models or combined model application may emerge as innovative solutions to reduce uncertainty. In a relevant recent study, three commonly used receptor modelling techniques (PCA, PMF and CMB) were tested for the same PM_{10} data set obtained from an industrial area in Spain (Viana, M. *et al.*, 2008). The comparisons of the results between the different models and validation against measurement data evidenced a good agreement regarding source identification, however larger differences were obtained regarding the quantification of source contributions. The analysis of the results suggested that the combined use of factor analysis techniques (PCA, PMF) to identify emission sources and the subsequent application of CMB for more detailed source apportionment and quantification of contributions would provide an appropriate methodology to overcome model limitations.

REVIEW OF SOURCE APPORTIONMENT USING MODELS IN EU COUNTRIES

In the previous section it was mentioned that the majority of the COST 633 countries that submitted the relevant questionnaires on methodologies used for source apportionment have reported the application of receptor models, as shown in the following table. Receptor modelling was used in 70% of the countries, whereas a combination of receptor and dispersion modelling was used in 20% of the countries and source apportionment based solely in dispersion modelling was reported for only one country, Austria, accounting for 10% of the countries questioned.

Table 1. Modelling tools used for source apportionment by different EU countries, according to the COST 633 questionnaire

Country	Modelling Methods
Austria	Dispersion model
Finland	Receptor modelling (PCA, MLR, MLF, SEM)
Germany	Dispersion model and Receptor modelling (PCA, MLR, PMF)
Greece	Receptor modelling (MR/APCS, CMB)
Italy	Dispersion model and Receptor modelling (PCA, PMF)
Netherlands	Receptor modelling (PCA, MLR)
Portugal	Receptor modelling (MLRA, PCA, MBA)
Spain	Receptor modelling (MLRA, PCA)
Sweden	Receptor modelling (PMF)
United Kingdom	Receptor modelling (PCA)

*PCA: Principal Component Analysis, MLR(A): Multivariate Linear Regression (Analysis), MLF: Maximum Likelihood Factor analysis, SEM: Structural Equation Modelling, PMF: Positive Matrix Factorisation, MR/APCS: Multiple Regression/regression on Absolute Principle Components, CMB: Chemical Mass Balance, MBA: Mass Balance Analysis

The use of dispersion models for source identification and quantification of corresponding contributions was significantly higher when natural sources of particulate matter were specifically addressed, according to the presentations from 10 EU countries which participated in the workshop on “Quantification of the contribution of natural sources to the ambient PM concentrations” in October 2006 in Ispra. Modelling was used in 90% of the cases, with the exception of the Netherlands, as the main focus of the relevant presentation was on sea-salt contribution, for which case the use of modelling tools is currently limited, but gradually growing. Fifty percent of the countries have used dispersion models, mainly Eulerian Chemical Transport Models, while 40% of the countries report the application of receptor models for source apportionment. In order to enhance the reliability of the methodology, 30% of the countries have applied back-trajectory analysis in combination with other modelling methods.

The increased use of dispersion models and of the combination of models for the quantification of natural contributions is also reflected in the technical reports submitted by EU countries in support of their application for postponement of attaining PM_{10} limit values. As the AQD allows for a 3-year extension of the attainment deadline in cases where exceedances can be explained due to contributions from natural sources, adverse climatic conditions and specific dispersion characteristics, the applicant EU countries had to develop a reliable methodology to: (a) confirm that a significant number of exceedances or high annual mean concentrations were due to natural sources, (b) to quantify the proportion of these exceedances and (c) to determine the extent to which the different natural sources were responsible by estimating the PM_{10} concentrations resulting from their relevant emissions.

At the time of preparation of this study, the EC has announced decisions for the time extension applications of 17 EU countries, including 289 air quality management zones. A demand for extending the period for attainment of the daily limit was expressed for the majority of the zones (287 zones), while a demand for extending the period for attainment of the annual limit regarded 230 zones. Nine of the applicant countries (53% of the total) considered transboundary pollution as the main cause for non-compliance, while 2 (Denmark and Austria) countries attributed a significant number of exceedances to winter-salting and sanding. Objections were raised by the EC for 96% of the zones applying for postponement of attaining the annual limit and for 86% regarding the daily limit. It is interesting to note that objections raised for 53% of the applicant countries (either referring to the annual or to the daily limit) were attributed to inadequate or incomplete source apportionment.

The analysis of the reports submitted by the countries complementary to the time extension applications suggests that receptor modelling was used to a smaller extent (29%) than dispersion models (41% Lagrangian, 59% Eulerian and 35% Gaussian) and trajectory models (41%). This can be explained on the basis of the transboundary contributions which represented in most countries a large percentage of natural contributions. The long-distance transport of particulate matter dictates the need to account for the physical and chemical processes governing pollutant transfer, while back-trajectories constitute an ideal first screening approach for identifying the origin of transported polluted air masses. Local scale modelling

(CFD software) was only applied by one country, as usually the impact of natural and transboundary sources in street canyons is less pronounced than the impact of local anthropogenic sources.

It is of great interest to note the high percentage of member countries (71%) that have applied a combination of modelling approaches for source apportionment. In several cases, Eulerian dispersion models were complemented by Lagrangian trajectory models to account for transboundary contributions, such as for Cyprus, Portugal and Spain (natural transboundary contributions), and Belgium and Austria (anthropogenic transboundary contributions). In Greece and Italy, Eulerian dispersion models have been used to account for transboundary transport of polluted air masses in combination with statistical receptor models for source attribution of both local/national and long-distance sources, while in Slovakia a Gaussian model was used for air quality assessment complemented by a Eulerian Chemical Transport Model (CTM) to assess transboundary contribution. Slovakia and Poland were the only countries to account for resuspension using the EPA emissions modelling approach, which requires input information on traffic characteristics, dust load on the road and street geometry.

Several countries have verified the model results against available measurements within the frame of the application, while the majority of the models used by the member countries for source apportionment are extensively validated in the literature. In some cases, such as for the United Kingdom, France and Portugal, model validation was explicitly described. The report submitted by the United Kingdom explains the use of a Volatile Correction Model to calibrate and validate the model results by applying appropriate scaling factors prior to the comparison with measurements. Portugal refers to the use of the "Standard Guide for Statistical Evaluation of Atmospheric Dispersion Model Performance" (ASTM, 2005) that has been consulted to validate the prognostic meteorological and air pollution TAPM modelling system. Finally, France applied the Eulerian CTM modelling system PREV'AIR to estimate transboundary and natural contributions, including on-line verification procedures.

Table 2. Modelling tools used for source apportionment by different EU countries for the purposes of preparing the time extension reports

Model type	Number of countries	% *
Lagrangian	7	41
Eulerian	10	59
Trajectory	7	41
Receptor	5	29
Gaussian	6	35
CFD	1	6
Combination of models	12	71

*Percentages do not add up to 100%, as many countries used more than one model type

CONCLUSIONS

The review of the reports submitted by the EU member countries to support their applications for postponement of the compliance deadline regarding PM_{10} limit values confirms the increased use of modelling tools for source apportionment in member states, which was also reflected in the presentations of the workshop on the "Quantification of the contribution of natural sources to the ambient PM concentrations". The majority of the countries applying for time extension have not applied a uniform methodology for source apportionment in all air quality management zones. It is therefore necessary to develop a standardised methodological framework for source apportionment using models including local, anthropogenic, natural and transboundary contributions, a range of pollutants and specific cases such as emissions due to resuspended road sand and salt. Specific attention should also be given to certain compounds that are yet not adequately quantified (biogenic secondary organic material and the nitrate component), to the apportionment of specific anthropogenic emission sources not sufficiently discriminated in many source apportionment studies (e.g. shipping emissions) and to the identification of biomass combustion sources.

The harmonised use of models for source apportionment suggested by the FAIRMODE SG will be complementary to the methodology suggested in the Guidance Document compiled by the EC Work-Group for Implementation and will include an assessment methodology for model evaluation. Particularly in the case of receptor statistical models, the solution does not guarantee physical reality and thus a validation procedure is essential to ensure reliable outcomes. For this purpose, statistical receptor models are preferably used in conjunction with dispersion models, especially to justify emission reduction measures on different source types. A standardised set of parameters and indices will be recommended to describe model quality in terms of accuracy, verification record and applicability to areas of specific interest (such as urban areas of complex topography) or for specific pollutants (such as organic aerosols). Guidelines will be also be provided in relation to the temporal and spatial resolution of both the models as well as of emission inventories and monitoring data. As for all modelling tools used for regulatory purposes, total model uncertainty has to be considered for models used in source apportionment, including model uncertainties, emission inventory uncertainties and uncertainties relating to meteorological variability during air pollution episodes.

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